Recent Advances in Development of Mercuric Iodide Radiation Detectors

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Abstract--**The performance of mercuric iodide radiation detectors has improved during the past two years to the point where they can be generally applied in instruments for the measurement and spectral analysis of nuclear radiation. This is due to systematic optimization of the processes to prepare the material and to fabricate the detectors. This paper will review these accomplishments in terms of spectral resolution, long-term stability and elevated temperature results. New detector arrangements and electronic systems, which have been developed to match the properties of the detectors, will be described.**

I. INTRODUCTION

Within the group of materials that are under active development as solid state radiation detectors with a wide dynamic range and operate at or near ambient temperatures, mercuric iodide has perhaps the most promising characteristics based on its theoretical physical and technological properties. The high values of the absorption coefficient, the photoelectric effect, the resistivity at room temperature, the resistance to damage by nuclear radiation of any kind and the compositional stability of the material during processing and crystal growth make mercuric iodide detectors the preferred candidates for a wide variety of applications. The practical realization of these superior properties in operating detectors, however, has required a long-term effort, which only recently has resulted in devices which have useful applications.

Detectors which are produced to measure and identify nuclear radiation have applications in the monitoring and safeguarding of stored nuclear materials, the detection of unusual, illegal or dangerous processing of these materials, the detection of illicit transportation, the characterization of nuclear wastes, nuclear medicine, environmental protection and remediation, and astrophysics experiments.

To be used for these purposes, the detectors are required to have a high sensitivity so that weak radiation intensities can be detected and measurement times can be short, but at the same time the detectors and the associated electronic systems need to be compact and rugged, especially if they are to be used in portable instruments.

In the case of mercuric iodide, this implies that the volume of the detectors needs to be maximized while at the same time the spectroscopic properties are optimized. This objective can be achieved by reducing the electronic defects in the material caused by impurities and native defects, and by increasing the structural quality of the material. The first of these problems can be solved by improvements in purification and adjustments of the stoichiometry. The second can be approached by more precise control of the crystal growth process and the optimization of the detector fabrication.

This paper describes the activities of the detector development group at Constellation Technology Corporation during the past two years. This activity has been directed towards implementing improvements in material preparation and detector fabrication. Results will be presented and discussed. In addition, some new developments in detector system fabrication and electronic systems are shown.

II. MATERIAL PREPARATION AND CRYSTAL GROWTH

The general methods used to synthesize mercuric iodide from aqueous solutions of mercuric chloride and potassium iodide, purify the resulting powder and grow the single crystals are still the same as described in [1]. Improvements in the material quality are the result of modifications and refinements in the procedures.

A. Synthesis and Purification

Minor changes have been made in several of the synthesis and purification steps on an experimental basis. Where these modifications had a distinct positive result, they have been made part of the standard procedures. As a result, the impurities in the material have been reduced to the lowest levels we have been able to determine. Two methods of analysis were used. Internally, we measured the concentrations of cations by ICP-AES. In addition, samples have been submitted to a commercial laboratory for independent evaluation of the results. This laboratory used the GDMS procedure which could determine both the cation and anion concentrations as well as total carbon.

Report Documentation Page

1. REPORT DATE **2001**	2. REPORT TYPE	3. DATES COVERED **00-00-2001 to 00-00-2001**

4. TITLE AND SUBTITLE **Recent Advances in Development of Mercuric Iodide Radiation Detectors**	5a. CONTRACT NUMBER
	5b. GRANT NUMBER
	5c. PROGRAM ELEMENT NUMBER
6. AUTHOR(S)	5d. PROJECT NUMBER
	5e. TASK NUMBER
	5f. WORK UNIT NUMBER
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) **Constellation Technology Corporation,7887 Bryan Dairy Road Suite 100,Largo,FL,33777**	8. PERFORMING ORGANIZATION REPORT NUMBER
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)	10. SPONSOR/MONITOR'S ACRONYM(S)
	11. SPONSOR/MONITOR'S REPORT NUMBER(S)

12. DISTRIBUTION/AVAILABILITY STATEMENT **Approved for public release; distribution unlimited**

13. SUPPLEMENTARY NOTES **The original document contains color images.**

14. ABSTRACT **see report**

15. SUBJECT TERMS

16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT	18. NUMBER OF PAGES **5**	19a. NAME OF RESPONSIBLE PERSON
a. REPORT **unclassified**	b. ABSTRACT **unclassified**	c. THIS PAGE **unclassified**			

The concentrations, in parts per million by weight, of 77 elements were measured. Table 1 presents a list of the elements detected in concentrations of parts per million. The concentrations of the other elements were in the low parts per billion range.

TABLE 1
IMPURITY ELEMENTS IN PPM

Element	GDMS	AES
Carbon	2.7	n/a
Sodium	0.1	2.2
Aluminum	0.4	0.1
Sulfur	0.3	n/a
Oxygen	2.0	n/a
Chlorine	32	n/a
Potassium	3.6	6.3
Bromine	1.7	n/a
Calcium	0.03	1.0
Iron	0.02	0.4
Silver	0.02	0.7
Barium	0.01	0.1
Lead	0.01	1.8

In general, there is reasonable agreement between the numbers obtained with the two different methods, and the total concentration of cation impurities is measured at approximately 10 ppm. The relatively large amounts of chlorine and bromine are thought to be introduced in the material through the use of mercuric chloride in the synthesis, and are an indication that the reaction is incomplete. The sulfur and oxygen impurities can be caused by sulfate and iodate anions in the reagents. The amount of total carbon detected is much lower than measured many years ago. One of the sources of carbon is thought to be the water used in the synthesis and other processes, and a major effort has been made to make our water purification system more efficient.

B. Crystal Growth

Single crystals of up to 500 grams can be grown routinely in closed ampoules by the physical vapor transport method as described in reference [1]. The established procedure is to nucleate a single seed on the pedestal in the bottom part of the ampoule and to continue the growth by slowly reducing the temperature in the lower section of the ampoule. This method gives generally satisfactory results, except that during the later part of growth, the surfaces of the crystals become unstable and new nucleation or voiding is created. The result is that only the central part of the crystal is of high enough structural quality to be processed into detectors and the yield is reduced. Since instabilities in crystal growth are generally caused by irregularities in the temperature profile of the crystal

growth furnaces, the temperature and cooling air flow controls have been upgraded during the past two years. Regrettably, these actions did not always have the desired effect. For this reason, an experimental program has been initiated recently whereby the temperature profiles in the furnaces and in the growing crystals will be modified in various ways so that the growth will be stable and regular until all the source material is converted into a single, high quality crystal. At this time it is too soon to report results from this effort.

III. DETECTOR FABRICATION AND TESTING

The process of cutting crystals into detector slices has been streamlined so that cutting losses are minimized. The standard sizes used to evaluate the material quality are 25 mm x 25 mm area and 2-3 mm thick or 10 mm x 10 mm area and 2-3 mm thick. Other dimensions can also be fabricated, for example 2 mm x 2 mm area and 10 mm long for use in medical probes. The preferred way of contacting is with sputtered Palladium. After contacting and coating with a polymer, the detectors are mounted on a small PC board with standard dimensions and the detector contact leads are connected to pins in this board. Figure 1 is a photo of this mounting. In this way it is easy to plug these essentially modular detector units into electronic systems for testing and for applications in instruments.
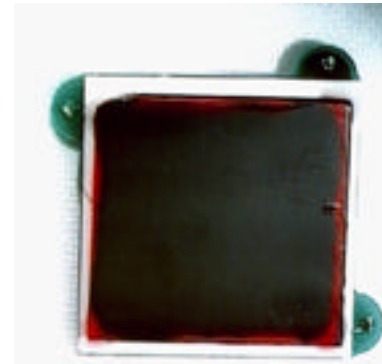

Figure 1. Finished detector 25 mm x 25 mm x 3mm mounted on PC board

The performance of the detectors is measured at a bias of approximately 1000 V/mm. Newly made detectors show, initially, relatively large leakage currents, so the bias is slowly increased over a period of two days to avoid damage to the preamplifier. This leakage current is thought to be caused by the removal of trapped charges. At this point, spectra can be acquired, but the optimal performance is not reached until the detector has been conditioned for several days. This conditioning time has been reduced during the past two years from an average of six days to two days. The effects of the conditioning are permanent and for actual use in an instrument the full bias can be applied in two to three minutes, and high quality spectra

can be collected within a short time. We think that the improvements in the detector performance as a result of conditioning may be due to the permanent removal of traps, for example iodine vacancies, from the detector body.

IV. DETECTOR PERFORMANCE

A variety of detector and contact structures, including planar x-ray and gamma ray, coplanar grid, pixillated and photosensitive detectors, has been evaluated in our laboratory and by other investigators. Results of these tests have been published in the past [2],[3] or will be presented at this workshop [4]. Some significant recent results will be shown in the following sections.

A. High Energy Gamma Ray Detectors

The spectrometer detectors fabricated to detect high energy gamma rays (400 – 1600 keV) are about 3 mm thick and have a resolution of 3% FWHM or better at the 661 keV energy of Cs-137. These detectors have a Minimal Detectable Activity (MDA) better than a 1 inch x 1 inch sodium iodide detector coupled with a photomultiplier tube [5]. Using specialized signal processing electronics, however, the resolution can be improved to less than 2%. An example is shown in Figure 2.
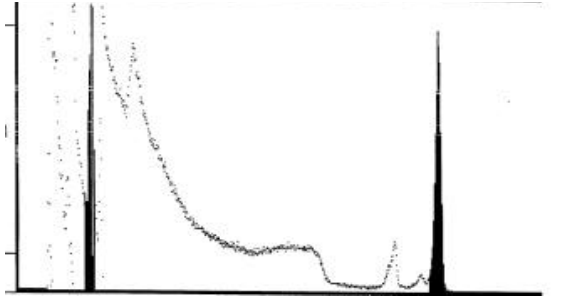

Figure 2. Cs-137 Spectrum of mercuric iodide detector (Courtesy R.Regal and P.Siffert, Eurorad and M.Hage Ali, PHASE-CNRS, Strasbourg, fr)

The resolution of this spectrum is approximately 1.4% FWHM. It is interesting to note that, at this resolution, the iodine escape peak becomes visible in addition to the normally present mercury escape peak at 580 keV. The noise level in the energy region of the main energy peak is very low, so that the Compton shelf is well defined. To obtain this spectrum, only about 10% of the incomplete charge pulses had to be rejected.

B. Low Energy Gamma Ray Detectors

For energies below 200 keV, the thickness of the detectors can be reduced while still maintaining a high efficiency because of the intrinsic properties of mercuric iodide. An example of this is shown in Figure 3 [6]. A 1 cm2 area and 2.7 mm thick mercuric iodide detector was subjected to the radiation from a weak U-235 source. The resolution of the spectrum was 5.7 keV FWHM at 186 keV

or 3.1%. The spectrum clearly separates the significant lines in the spectrum, including the uranium and thorium fluorescence lines caused by self-absorption in the material. The spectrum is of sufficient quality to be amenable to a variety of software analysis systems.
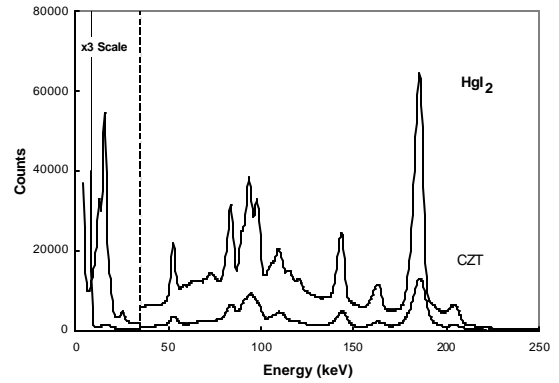

Figure 3. Spectra of U-235 measured with a Mercuric Iodide detector and a CZT detector.

For comparison, the same spectrum was measured under identical conditions using a 1 cm2 area and 5 mm thick coplanar-grid CZT detector. The same pulse-processing electronics were used for both detectors, although the settings were optimized for each independently. The intensity of the mercuric iodide spectrum is about 4 times higher, reflecting the higher intrinsic efficiency, and the spectral peaks are better defined, which improves the evaluation of the nuclear material.

C. Stacked Detectors

In order to improve the overall efficiency of a mercuric iodide detector system, four detectors have been combined by placing them on top of each other in a stack. This arrangement was designed to detect the high energy gamma rays (3-11 MeV) which are generated in neutron activation analysis. The individual detectors use the same bias supply, and the signals are routed to a common preamplifier and electronic signal processing system [7]. The total active volume of the stack was approximately 6 cm3 . We prefer the stacked arrangement over a flat array because, in a stack, the individual detectors are in very close proximity to each other. As a result, the probability is high that escape peaks and Compton-scattered photons exiting from one detector are captured in adjacent detectors. Figure 4 shows a Cesium-137 spectrum of a stack.
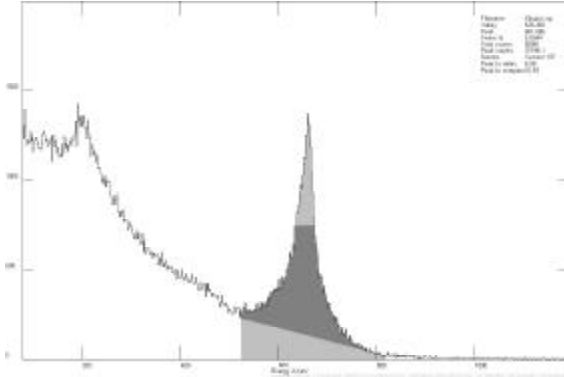
Fig. 4. Spectrum of Cs-137 measured with a 4-element stack.

The resolution of the individual detectors was about 6%, but the resolution of the stack is 5.6% FWHM. The mercury escape peak has disappeared from the spectrum, and the full energy peak efficiency has increased compared with that of the individual detectors. Several of these stacks can be combined in an array to increase the efficiency of the system even further. In addition, the signals of the individual detectors can be processed separately and can be summed using a timing system. In this way, coincidence of the different signals can be monitored and noise caused by scattering events or charged particle interactions can be rejected. This feature is especially important for gamma ray astronomy, since a coincidence shield will become redundant and the weight available for detector systems can be used exclusively for active gamma ray measurements.

V. TRANSPORT PROPERTIES OF DETECTORS

The improvements in the spectral performance have been substantiated by measurements of the transport properties of the charge carriers for a series of detectors. In good detectors, the values of the mu-tau product for electrons is approximately 1×10^{-4} cm2/V and for the holes approximately 5×10^{-5} cm2/V. One of the explanations for these results can be the careful adjustment of the mercury and iodine balance in the material so that hole trapping is minimized, as suggested by Sharma, Pal and Acharya [8].

The values of the mobilities of electrons and holes for many detectors vary between 60-100 cm2/Vsec for electrons and 2-4 cm2/Vsec for holes respectively. The low value of the hole mobility at this time puts a limitation on the thickness of detectors to be used for spectroscopy, and thereby on their efficiency, especially at energies higher than 500 keV. The average transit time for a hole through a 3mm detector is approximately 10 microseconds. This increases the trapping statistics and necessitates long shaping times for full charge collection, thereby increasing the electronic noise. Nevertheless, the performance of mercuric iodide detectors, in terms of spectral resolution and efficiency, compares favorably with that of other room-temperature detectors without having to resort to complicated contact structures or elaborate electronic compensation systems. The reasons for this are the large active volumes available, the intrinsic high efficiency of the material and the low leakage currents of the detectors.

It is the considered opinion within our group that the mobilities in a single crystalline material are determined primarily, but not exclusively, by the structural perfection and homogeneity of the material, so that phonon scattering caused by extended defects is minimized. At the same time, the trapping times are primarily determined by the concentration and nature of atomically distributed defects such as vacancies and interstitials generated by impurities and native defects. It is for these reasons that we take a two-pronged approach to the improvement of the detector performance. One program has been initiated to modify the temperature profiles in the crystal growth furnaces to increase the quality and yield of crystalline material with high structural perfection. A second effort is directed towards increased purification and adjustment of the mercury/iodine balance in the material. Results of these experiments are expected to be available in the near future.

VI. IMPROVEMENTS IN ELECTRONICS

When thick detectors are used for high resolution spectroscopy and a semi-gaussian shaping amplifier is used to process the signals from the detector, it is necessary to apply shaping times as long as 24 microseconds to collect the charges because of the low values of the mobilities and the trapping times of the holes. This restricts the count rate to several thousand counts per second. In addition, the long shaping times add between 3-5 keV electronic noise, which has a significant effect on the spectral resolutions at energies less than 150 keV. To overcome this limitation in count rates and to minimize the ballistic deficit and the electronic noise, a gated integrator circuit has been designed which can be adjusted by software commands to match the electronic properties and the applications of the detectors. Using the gated integrator circuit, the count rate of the detector can be increased to approximately twenty thousand counts per second without loss of spectral resolution and with minimal pile-up of signals.

The programmable gated integrator amplifier has been implemented in a handheld spectrometry system called the Micromax. This unit has dimensions of 14 mm x 7 mm x 3 mm and its weight is less than 250 gram. Figure 5 shows a photograph of this assembly.

Fig. 5. Photograph of the Micromax.

The Micromax performs a series of functions needed to acquire spectra. It can provide up to 3000 V to a detector module and up to +12 V to a preamplifier, so that it can be used with a variety of detector systems. The Micromax generates 1024 channel spectral data which can be downloaded through a serial port or can be stored internally in a non-volatile memory. The Micromax is powered by two internal AA batteries or by 7-12 VDC external power.

VII. SUMMARY

The development of mercuric iodide detectors during the past two years has given rise to gradual improvements in detector performance, due to continual optimization of established procedures. As a result, a variety of detector structures with high efficiency and spectral resolution can be fabricated, which have been incorporated into portable and stationary functional instruments.

The factors which limit increased detector thickness and efficiency are the mobility and the trapping times of the holes. These limitations can be eliminated by growing single crystalline material of higher structural quality and by reducing the density of critical trapping centers through purification and adjustment of the stoichiometry. Experimentation in these areas will continue, combined with the routine monitoring of the electronic charge properties.

The introduction of a gated integrator system to process the signals from the preamplifier has increased the count rate of the detectors and has reduced the ballistic deficit. In addition, the electronic noise levels have been reduced so that the spectral resolution at lower gamma ray energies has improved.

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